Optical nonlinearities induced in a magnetic liquid by an alternating magnetic field

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The spectral composition of the oscillations in the orientation of particles in a magnetic liquid in a large-amplitude alternating field has been found. The results show that the superparamagnetism of the particles gives rise to some characteristic structural features in the intensity of polarized light transmitted through the magnetic liquid. The structural features consist of minima in the field dependence of harmonic amplitudes.

Birefringence measurements have been used successfully[1-3] to study magnetic liquids, i.e., suspensions of single-domain ferromagnetic particles. The optical anisotropy of these media is extremely high: In magnetite magnetic liquids with a solid-phase concentration of 3–5% by volume, for example, the Cotton-Mouton constant is larger by a factor of 10^6–10^7 than that for molecular liquids.[4] Such a dramatic response to an applied field is of course due to the significant value of the magnetic moment μ of an individual ferromagnetic particle. Specifically, if the size of the particle is ~ 100 Å, we have $\mu \sim 10^{-15}$ erg/G, i.e., about 10^5 magnetons. An estimate of the value of the parameter $\xi = \mu H / k_B T$, which determines the orientation of the particle in a magnetic field H at a temperature T, yields $\sim 10^{-2}\, H$ for $T = 300$ K. In other words, ξ reaches a value of unity even in moderate fields. Beginning at $H \gtrsim 100$ Oe, the field dependence of the orientational effects in magnetic liquids obviously becomes very nonlinear. In this letter we are reporting a study of how these nonlinearities influence the dynamics of the birefringence in magnetic liquids.

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We consider a dilute suspension of single-domain ferromagnetic particles with a volume $V \sim 10^{-18}$ cm^3. We assign the particle the shape of a prolate ellipsoid of revolution, assuming that the anisometry is sufficient to produce a uniaxial magnetic anisotropy with an easy-magnetization direction along the principal axis of the ellipsoid. We characterize the orientation of this axis by the unit vector $\bar{\nu}$. Using typical values of the magnetic-anisotropy constant, $K \sim (1\text{–}5) \times 10^5$ erg/cm^3, and estimating the ratio of the anisotropy energy of a particle to its thermal energy at $T = 300$ K, we find $\sigma = KV/k_B T \lesssim 5$, indicating an intense orientational diffusion of the magnetic moment of the particle (a superparamagnetism).[5] In superparamagnetic particles with $\sigma < 5$, the response time of the magnetic moment, τ_0, does not exceed 10^{-8} s (Ref. 5), so that in fields $H(t)$ with frequencies $\omega\tau_0 \ll 1$ the orientation of the vector μ can be assumed to be at equilibrium.

In a linearly polarized field, the optical anisotropy of the suspension, Δn, is proportional to the quantity[6] $S = S_{ik} h_i h_k$, where $\mathbf{h}$ is a unit vector along the field direction, and $S_{ik} = 3/2(\langle \nu_i \nu_k \rangle - \delta_{ik}/3)$ is a macroscopic orientation tensor, determined by an average over the ensemble of particles. In the case of superparamagnetic particles in a low-frequency field, the equation of motion of the parameter S is

$$\tau \dot{S} + S = (2/15)\, \sigma \mathcal{L}(\xi). \tag{1}$$

Here $\mathscr{L}(x) = 1 - 3L(x)/x$, where $L(x)$ is a Lagrangian. The relaxation time $\tau = A\eta V/k_B T$ determines the dispersion of S (and thus Δn)—this is the Debye time of the rotational diffusion of a particle suspended in a liquid with a viscosity η. The form factor A is equal to unity for a sphere and increases with increasing anisometry. Even in a low-viscosity liquid we would have $\tau \geqslant 10^{-6}$ s, so the condition we are adopting here, $\omega\tau_0 \ll 1$, does not impose any stringent limitation on the value of $\omega\tau$.

Equation (1) differs from the equation of motion of the orientation tensor of nonpolar molecules in an electric field[7] only in the form of the right side. Equation (1) contains a saturating function of the field, not simply a quadratic function as in the case of electropolarization. The effect is to impart a substantial anharmonicity to the driving force.

Let us examine the frequency spectrum of the steady-state oscillations in $S(t)$ in a field $H = H_0 \cos \omega t$. Setting $\xi = \xi_0 \cos \omega t$, where $\xi_0 = \mu H_0 / k_B T$, in (1) and expanding the unknown quantity in a harmonic series,

$$S(t) = \sigma \sum_{l=0} s_{2l\omega} \cos(2l\omega t - \varphi_l), \tag{2}$$

we find from (1) and (2) a system of algebraic equations for the spectral amplitudes $s_{2l\omega}$ $(\xi_0, \omega\tau)$. From (1) and (2) we easily find the dispersion law $s_{2l\omega} \propto 1\sqrt{1 + (2l\omega\tau)^2}$ and the asymptotic behavior $s_{2l\omega} \propto \xi_0^{2l}$ in weak fields. The spectrum of function (2) has been constructed numerically for arbitrary values of ξ_0. Figure 1 shows the results calculated for the amplitudes of the first few oscillation modes.

The most accessible method for experimental determination of the dependence $S(t)$ is to measure the birefringence. Let us examine the influence of nonlinear effects on the frequency spectrum of the intensity of light which has been transmitted through

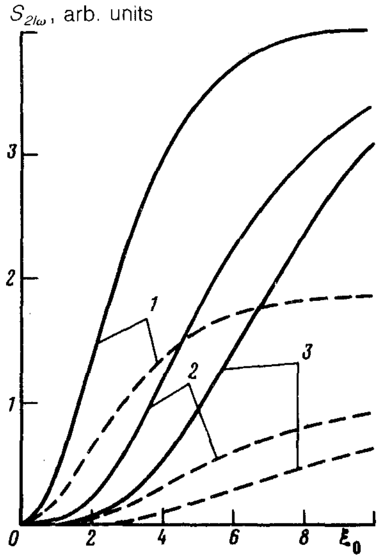

FIG. 1. Dependence of the frequency spectrum of the orientation parameter on the amplitude of the applied field. 1—$s_{2\omega}$; 2—$2s_{4\omega}$; 3—$4s_{6\omega}$ for $\omega\tau = 0.1$ (solid lines) and for $\omega\tau = 1.0$ (dashed lines).

a layer of a magnetic liquid. We assume the standard experimental geometry: The polarizer and the analyzer are crossed, and the field makes an angle of 45° with the axes of the polarizer and the analyzer. Under these conditions the relative intensity of the transmitted light is described by $I = \sin^2(\delta/2)$, where δ is a phase shift proportional to Δn. For a dilute suspension, with $\delta \ll 1$, we would have $I \approx \delta^2/4 \propto S^2$. Using

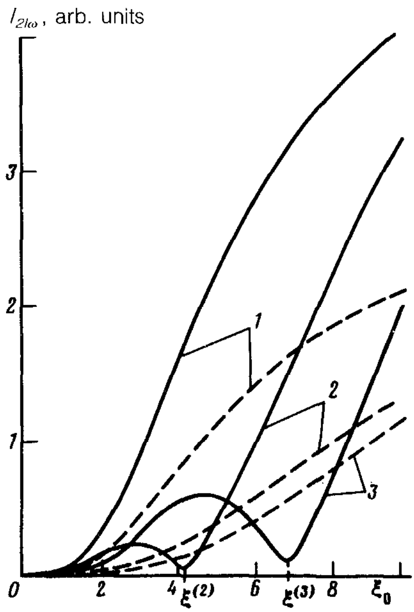

FIG. 2. Frequency spectrum of the intensity of the polarized light, $I(t)$, versus the amplitude of the applied field. 1—$I_{2\omega}$; 2—$3I_{4\omega}$; 3—$8I_{6\omega}$ for $\omega\tau = 0.1$ (solid lines) and for $\omega\tau = 1.0$ (dashed lines).

TABLE I.

l	2	3	4
$\xi^{(l)}$	4,2	6,8	9,4

this relation to transform the spectrum $S(t)$ found above into an $I(t)$ spectrum, we find the pattern shown in Fig. 2 (the constant component is omitted from this figure, as it has been from Fig. 1). We see that at $\omega\tau \ll 1$ the behavior of the spectral amplitudes at all $l > 1$ is obviously nonmonotonic: We see a minimum, whose position $\xi^{(l)}$ shifts toward a higher amplitude of the applied field with increasing index of the harmonic. The l dependence of $\xi^{(l)}$ was found numerically. The calculations for $\omega\tau \leqslant 0.1$ are shown in Table I. As the field frequency is increased, the positions of the minima on the $I_{2l\omega}$ (ξ_0) curves vary only slightly, but their depths decrease noticeably. At the same time, the amplitudes $I_{2l\omega}$ themselves decrease. In the model which we are discussing here, this decrease would be explained by the absence of orientation oscillations in the limit $\omega\tau \to \infty$.

To test this theory, we will use the results of Ref. 8. Skibin[8] studied the modulation of light by an optical cell filled with a ferromagnetic colloid with a magnetite concentration of 0.15% by volume. A nonmonotonic dependence $I_{2l\omega}(H0)$ was seen experimentally there for the first time. Here is a comparison of the ratios from Ref. 8 of the field amplitudes $H^{(l)}$ corresponding to the minima of the spectral amplitudes of the intensities with the numbers from Table I:

$$\xi^{(3)}/\xi^{(2)}\Big|_{\text{expt}} \approx 1.4 \qquad \xi^{(3)}/\xi^{(2)}\Big|_{\text{theo}} = 1.6$$

$$\xi^{(4)}/\xi^{(2)}\Big|_{\text{expt}} \approx 1.9 \qquad \xi^{(4)}/\xi^{(2)}\Big|_{\text{theo}} = 2.2 .$$

The quantitative agreement between theory and experiment is satisfactory. To determine the size of the ferromagnetic particles, we use the value $H^{(2)} \approx 500$ Oe given in Ref. 8, and we calculate the ratio $\xi^{(2)}_{\text{theo}}/H^{(2)}$. According to our model, this ratio would be equal to $\mu/k_B T$. Assuming $\mu = MV$ for the single-domain particles, where $M = 480$ G is the magnetization of magnetite, we find the estimate $V = k_B T\xi^{(2)}/MH^{(2)} \approx 6.8 \times 10^{-19}$ cm^3 at room temperature. In terms of a linear dimension, this would be ≈ 110 Å. This figure correlates well with the available data from electron microscopy of magnetite magnetic liquids.

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